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Source Category Survey
Uranium Refining Industry

Radian Corp.
Durham, NC

Prepared for

Environmental Protection Agency
Research Triangle Park, NC

May 80

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16. ABSTRACT

The purpose of this source category survey was to determine if new source performance standards (NSPS) should be developed for the uranium refining industry. Information on the uranium milling process, the uranium conversion process, industry growth, and industry emissions was collected. The U.S. Nuclear Regulatory Commission (NRC) and the U.S. Environmental Protection Agency Office of Radiation Programs were contacted to determine the existing and proposed regulations applicable to this industry. It was concluded that radionuclide emissions are regulated by the NRC under the Uranium Fuel Cycle Standard (40 CFR Part 190) and that all significant particulate emissions of uranium mills will be assessed (and regulated; if warranted) under the current project for development of NSPS for the metallic minerals industry. Based upon this information it was recommended that NSPS not be developed for the uranium refining industry.

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SOURCE CATEGORY SURVEY:
URANIUM REFINING INDUSTRY

1.0 BACKGROUND

The purpose of this report was to determine if new source performance standards (NSPS) should be developed for the uranium refining industry. Based on a 1976 report on uranium mining and milling by the Battelle Columbus Laboratories, the uranium refining industry was placed on the priority list for NSPS under the Clean Air Act Amendments of 1977. The Batelle report established domestic uranium milling capacity at 29,100 metric tons per day (MT/day) (32,000 tons per day (TPD)) or 10.6 million metric tons per year (MT/yr) (11.7 million tons per year (TPY)).[26] Later, uranium milling capacity for 1980 was predicted to be 25.4 million MT/yr (27.9 million TPY) ore. According to an estimate by the Argonne National Laboratories, a reduction of approximately 4,270 MT/yr (4,700 TPY) particulate emissions would result from a new source performance standard.[18]

The Uranium Refining source category was defined by the Office of Air Quality Planning and Standards (OAQPS) to include uranium mills and conversion facilities. Production of uranium concentrate (yellowcake) by nonconventional methods (in-situ leaching, copper leaching, by-production of wet-process phosphoric acid) was to be considered as an overview only. The Source Category Survey Report was to include all significant pollutants, not just particulates.

The survey included some interaction with the Office of Radiation Programs (ORP) of the Environmental Protection Agency (EPA) and the Nuclear Regulatory Commission (NRC). Regulatory authority and jurisdiction were documented since radionuclides were to be considered in the source category.

2.0 URANIUM MILLS

Uranium milling is the processing of uranium-bearing ore and production of uranium concentrate (yellowcake). The growth of uranium milling has

been established in the recent "Source Category Survey Report on Uranium and Vanadium Ore Processing."[9] Uranium concentrate production has increased 45 percent in the last 2 years as illustrated in Figure 1.[12,28,29]

The majority of uranium concentrate is produced in the Wyoming Basins and Colorado Plateau regions of the western states, as noted in Figure 2.[12,28,29] There are, however, a few small facilities, mainly in Florida, that produce concentrate as a byproduct of the phosphoric acid industry. An additional small amount of uranium concentrate is derived from in-situ mining and copper heap dump leaching. These nonconventional sources of concentrate contribute only five percent of domestic uranium concentrate production.[12,29]

2.1 Processes[5,6,7,9,13,19,26,27,30,31,32,33,35]

In general, two approaches are used in uranium ore processing: alkaline leaching (Figure 3) and acid leaching (Figure 4).

2.1.1 Alkaline Leach Process.

2.1.1.1 Leaching of ore. Alkaline leaching is normally done for ores of lime contents greater than 12 percent. The leaching solution is a mixture of sodium carbonate and sodium bicarbonate (or the ammonium compounds). This solution is fed to the grinders and used as the wetting agent in reducing particle size to approximately 200 mesh. The finer particle size is necessary since the leach solution cannot leach across grain boundries. The ground pulp is heated and fed to air-agitated Pachuca tanks. Leaching is done at 82-135°C (180-275°F) and takes from 5 to 96 hours. Oxidants used in alkaline leaching include air, copper, ammonia, and permanganate.

2.1.1.2 Thickening of leach solution. Alkaline leaching is selective for uranium. Thus, the slurries need only have the solids removed. This is done by vacuum drum filtration. Simple countercurrent decantation is used for desanding when "resin-in-pulp" (RIP) is used for purification. The residue is sent to the tailings pond.

2.1.1.3 Precipitation and dewatering. Sodium hydroxide is used to precipitate the uranium concentrate (uranium oxides and sodium uranates). The concentrate is filtered and dried. The filtrate is recarbonated and recycled to grinding or filtration.

2.1.2 Acid Leach Process.

2.1.2.1 Leaching of ore. Acid leaching is used for ores of lime content less than 12 percent. The most common acid used in uranium ore leaching is sulfuric acid. Leaching is done in open, agitated tanks. Oxidants (sodium chlorate or manganese dioxide) are added if there is insufficient ferric ion in the ore. Since sulfuric acid is a more effective leaching agent, the ore need only be ground to about 28 mesh. But, due to the increased leaching effectiveness, purification steps must be added to the process to eliminate other minerals that may have been leached into solution.

2.1.2.2 Thickening of leach solution. Acid leach mills generally use countercurrent decantation (CCD) to eliminate sand fines. This serves a two-fold purpose: (1) elimination of solids and (2) washing of fines to remove leach solution.

2.1.2.3 Purification. The leach solution is purified by ion exchange, solvent extraction, or Eluex processes (a combination of ion exchange and solvent extraction). Solvent extraction has become more popular since it is operated continuously and provides a pure product.

Ion exchange (IX) uses strong or intermediate base anionic-type resins. Strip solutions are normally composed of a chloride, nitrate, bicarbonate or ammonium sulfate-sulfuric acid combination. There are four types of ion exchange processes:

(1) fixed-bed type, with stationary resin-packed columns;

(2) moving-bed column type, where resin is transferred between stationary columns;

(3) continuous resin-in-pulp (RIP), where resin and solution flow countercurrently (agitation is by forced air); and

(4) basket RIP, where solutions flow in and out of tanks and the resin is held in agitated baskets.

Solvent extraction (SX) uses an organic phase extractant to withdraw the uranium from the leach solution. An aqueous phase extracts the uranium from the organic phase in a purified, concentrated state. There are two solvent extraction processes used in uranium milling:

(1) Dapex process: the extractant is an alkyl phosphoric acid (di(2-ethylhexyl) phosphoric acid (EHPA) as a 4 percent solution in kerosene). The modifier is usually tributyl phosphate (TBP), but long chain alcohols, such as isodecanol, are also used. The stripping solution, which is recycled, is sodium carbonate.

(2) Amex process: the extractant is an amine (6 percent almine-336 in kerosene), modified with isodecanol. Stripping is done with an ammonium sulfate solution maintained at a pH 4.0 to 4.3 to eliminate sodium impurities.

2.1.2.4 Precipitation and dewatering. Uranium concentrate or yellowcake is precipitated from the purified solution by addition of ammonia (other possible chemical additives are sodium hydroxide, hydrogen peroxide and magnesia). Before being fed to the driers, the yellowcake is partially dewatered by filtration or centrifugation.

2.1.3 Drying. Yellowcake drying is commonly done in a multiple hearth furnace. In some cases, such as excess sulfate in the product, the dryer is operated at a temperature sufficiently high to drive off sulfates as SO_3. This procedure often causes transformation of uranates to uranium oxides, along with the release of ammonia.

2.2 Emissions

2.2.1 Particulates. The major airborne emissions resulting from uranium ore processing are particulate matter. Particulates are emitted from crushing, grinding, and ore handling operations, in addition to drying and packaging of yellowcake. Particulate emissions from these sources will be assessed and regulated as warranted under the NSPS being developed for the metallic minerals industry.

2.2.2 Radionuclides. Uranium ore processing facilities are licensed by the NRC or by those States that have agreed with the NRC to perform as the regulatory authority (i.e., agreement States). The NRC is charged with enforcement of radionuclide emission standards, as established by ORP in the Uranium Fuel Cycle Standard (40 CFR Part 190). Radon emissions are not regulated under this standard since dosages are difficult to determine and emission control techniques are not known. Emission

regulations of agreement States must be as restrictive as the Uranium Fuel Cycle Standard.

Radionuclide emissions from milling operations include radon gas from ore handling and yellowcake dust from drying and packaging. Uranium mills are generally located at uranium mine sites. Based on this, the radon source represented by the mine is much larger than that for the mill. Also, radon is mainly emitted for dry uranium ore and exposure to dry ore in the mill is limited to ore handling and crushing. As a result, worker exposure to radon in the milling operation is considered minimal.

Yellowcake dust is minimally 90 percent U_3O_8 and would be considered a radionuclide. The emission of yellowcake dust would, therefore, be more tightly controlled as a radionuclide under the Uranium Fuel Cycle Standard than as a particulate under the NSPS being developed for the metallic minerals industry.

2.2.3 Other Emissions. Nonradiological emissions include hydrocarbons, SO_2 and acid mist (less than 8.2 MT/yr (9 TPY))[23], NO_x (less than 1.8 MT/yr (2 TPY))[23], and ammonia (trace amounts from yellowcake precipitation and drying).[21,22,23] Hydrocarbons are, by far, the largest of these emissions, with estimates ranging from 0.5 to 82 kilograms per day (1 to 180 pounds per day). As a worst case estimate, less than 30 MT/yr (33 TPY) of hydrocarbons per plant are emitted, mainly as kerosene, from solvent extraction purification operations.[8,26,27] These examples of nonradiological emissions from uranium mills, compiled from four NRC Final Environmental Statements, are detailed in Table 2.

3.0 URANIUM CONVERSION FACILITIES[1,6,17,30,31,32,33]

Uranium conversion facilities process uranium concentrate from milling operations to uranium hexafluoride (UF_6). Uranium hexafluoride is required for uranium isotope separation in gaseous diffusion plants.

There are only two existing conversion facilities in the United States: Allied Chemical Corporation's Metropolis (Illinois) Works (using a dry hydrofluor process) and Kerr-McGee Nuclear Corporation's Sequoyah (Oklahoma) plant (using a government-designed solvent extraction process). Production capacity could increase in the next 5 years by, at most, one new plant, or by expansion of an existing facility.

3.1 Dry Hydrofluor Process.

A block diagram of the dry hydrofluor process is shown in Figure 5. In this proprietary process, uranium concentrate is fed to a series of fluid beds where it is converted to uranium dioxide (UO_2). Through countercurrent contacting with anhydrous hydrofluoric acid, UO_2 is converted to uranium tetrafluoride (UF_4). Uranium hexafluoride is formed from UF_4 through contacting with fluorine in a series of fluorination towers. The gaseous UF_6 product is filtered and condensed to its solid state for packaging prior to shipment to diffusion plants.

3.2 Solvent Extraction Process.

As noted in Figure 6, the solvent extraction process differs from the dry hydrofluor process in that uranium concentrate is initially digested in hot nitric acid. Solvent extraction is then used for purification, with tributyl phosphate in hexane as the organic phase and water as the second extractant. The resulting uranyl nitrate is dehydrated and denitrated to uranium trioxide using heaters. Reduction to uranium dioxide is done by countercurrent contacting with dissociated ammonia at 593°C (1100°F) in a two-stage fluid bed. The further conversion to UF_6 is as in the dry hydrofluor process.

3.3 Emissions.

Both conversion facilities are located in nonagreement states. Thus, the NRC has licensing authority and regulatory jurisdiction of each plant.

3.3.1 Radionuclides. Particulate emissions from conversion facilities are, as for yellowcake, considered radiological. The radionuclides are regulated by the NRC under the Uranium Fuel Cycle Standard.

3.3.2 Fluorides. Combined fluoride emissions at each facility are currently within air quality standards. Since Oklahoma and Illinois have no fluoride air quality standards, these conversion facilities use the standard of the State of Washington (0.5 $\mu g/m^3$) as a reference. These emissions are controlled using scrubber systems (water, caustic, KOH) to meet fluoride standards and to guard against radionuclide emissions. The worst case emission has been noted as 0.22 $\mu g/m^3$ fluoride at the

fenceline of Allied's Metropolis Works.

3.3.3 Hydrocarbons. Hydrocarbon emissions result from solvent extraction purification at the Sequoyah facility. These emissions are as hexane and, as an uncontrolled worst-case estimate, amount to 168 MT/yr (185 TPY) or 5.33 grams per second (g/sec). This emission rate does meet the Oklahoma hydrocarbon emission standard (15.88 g/sec). According to the State of Oklahoma, the emissions of hydrocarbons from this facility are not significant.

4.0 NONCONVENTIONAL URANIUM PRODUCTION

Uranium production by nonconventional methods includes in-situ leaching, copper heap pile leaching, and byproduction of wet-process phosphoric acid. These methods combine to represent only five percent of domestic uranium production. Although no emission data are available, emissions would probably be lower than those of mills due to the magnitude of operations.

5.0 CONCLUSIONS AND RECOMMENDATIONS

Based upon this information, it was concluded that:

(1) Radionuclide emissions are regulated by the NRC) under the Uranium Fuel Cycle Standard (40 CFR Part 190). Particulate matter in the product end of the milling process has a high radionuclide content and is more tightly regulated as a radionuclide than one would expect for a particulate NSPS.

(2) All significant particulate emissions of uranium mills will be assessed (and regulated; if warranted) under the current project for development of NSPS for the metallic minerals industry.

(3) Other emissions are minor and are currently meeting air quality standards.

It was, therefore, recommended that NSPS not be developed for the uranium refining industry.

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TABLE 1A. U.S. URANIUM PRODUCTION MILLS
OPERATING AS OF JANUARY 1, 1979[29]

Conventional Mills	Plant Location
The Anaconda Copper Company	Grants, New Mexico
Atlas Minerals Corporation	Moab, Utah
Bear Creek Uranium Company	Powder River Basin, Wyoming
Conoco-Pioneer Nuclear	Falls City, Texas
Cotter Corporation	Canon City, Colorado
Dawn Mining Company	Ford, Washington
Exxon Minerals Company, USA	Powder River Basin, Wyoming
Federal American Partners	Gas Hills, Wyoming
Kerr-McGee Nuclear Corporation	Grants, New Mexico
Pathfinder Mines Corporation	Gas Hills, Wyoming
Pathfinder Mines Corporation	Shirley Basin, Wyoming
Petrotomics Company	Shirley Basin, Wyoming
Rio Algom Corporation	LaSal, Utah
Sohio Natural Resources Company	Cebolleta, New Mexico
Union Carbide Corporation	Uravan, Colorado
Union Carbide Corporation	Natrona County, Wyoming
United Nuclear Corporation	Church Rock, New Mexico
United Nuclear--Homestake Partners	Grants, New Mexico
Western Nuclear, Inc.	Jeffrey City, Wyoming
Western Nuclear, Inc.	Wellpinit, Washington
Solution Mining Operations	**Plant Location**
IEC Corporation	Pawnee & Ray Point, Texas
Mobil Oil Corporation	Bruni, Texas
Union Carbide Corporation	Palagana, Texas
United States Steel Corporation	George West, Texas
U.S. Steel-Niagara Mohawk	George West, Texas
Wyoming Mineral Corporation	Bruni, Texas
Wyoming Mineral Corporation	Ray Point, Texas
Wyoming Mineral Corporation	Irigaray Wyoming
Heap Leaching: Dumps, Tailings or Copper Dumps	**Plant Location**
Durita Development Corporation	Naturita, Colorado
Solution Engineering, Inc.	Falls City, Texas
Union Carbide Corporation	Maybell, Colorado
Wyoming Mineral Corporation	Bingham Canyon, Utah

TABLE 1B. U.S. URANIUM PRODUCTION MILLS
CURRENT PROPOSED PROJECTS[9,13,20,27]

Conventional Mills	Plant Location
American Nuclear Corporation	Gas Hills, WY
Bokum Resources Corporation	Marquez, NM
Chevron Oil	Panna Maria, TX
Conoco	Crownpoint, NM
Cyprus Mines Corp./WMC	Canon City, CO
Energy Fuels Nuclear, Inc.	Blanding, UT
Gulf Mineral Resources	Mt. Taylor, NM
Homestake Mining Co.	Gunnison, CO
Kerr-McGee Nuclear Corp.	Casper, WY
Mineral Energy	Temple Mountain, UT
Minerals Exploration	Sweetwater, WY
Mobil Oil Company	Crownpoint, NM
Phillips Uranium	Nose Rock, NM
Pioneer Nuclear, Inc.	McKinley Co., NM
Pioneer-Uravan	Slick Rock, CO
Plateau Resources, Ltd.	Shootering Canyon, UT
Portland General Electric/ Martin-Trost Assoc.	Miracle Mine, CA
TVA	Edgemont, SD
Union Oil	Shirley Basin, WY
United Nuclear Corporation	Morton Ranch, WY
Solution Mining Operations	**Plant Location**
Cleveland Cliffs Iron Co.	Pumpkin Buttes, WY
Ogle Petroleum	Bison Basin, WY
Rocky Mountain Energy Company	Casper, WY
Texura Corporation	Hobson, TX
Wyoming Mineral Corporation	Buffalo, WY
Phosphoric Acid Byproduct	**Plant Location**
Gardinier, Inc.	Tampa, FL
IMCC	Mulberry, FL
Heap Leaching: Dumps, Tailings, or Copper Dumps	**Plant Location**
Anamax	Tucson, AZ
Brush Wellman, Inc.	Delta, UT
Phelps Dodge	Bisbee, UT

TABLE 2. ESTIMATES OF EMISSIONS FROM URANIUM MILLS[8,15,22,23,24]

Uranium Project	Capacity[a] MT/day (TPD)	Emissions Rates[b] kg/hr(lbs/hr)	MT/yr(TPY)
Bear Creek	1270(1400)		
Leaching $SO_2 + H_2SO_4$		0.009(0.020)	0.08(0.09)
Chlorine		0.010(0.022)	0.09(0.10)
Solvent Extraction Hydrocarbons		0.014(0.030)	0.12(0.13)
Drying SO_2		0.45(1)	4.0(4.4)
NO_x		0.014(0.030)	0.12(0.13
Tailings Hydrocarbons		3.3(7.2)	28.6(31.5)
Shootering Canyon	718(790)		
Solvent Extraction Hydrocarbons		0.94(2.1)	8.4(9.2)
Highland	2730(3000)		
Solvent Extraction Hydrocarbons		0.18(0.04)	0.16(0.18)
White Mesa	1820(2000)		
Leaching $SO_2 + H_2SO_4$		0.023(0.05)	0.18(0.20)
Solvent Extraction Hydrocarbons		0.045(0.1)	0.37(0.41)
Drying SO_2		0.91(2)	7.5(8.2)
NO_x		0.23(0.5)	1.8(2.0)

[a]MT/day is metric tons per day; TPD is tons per day.

[b]kg/hr is kilograms per hour; lbs/hr is pounds per hour; MT/yr is metric tons per year; TPY is tons per year.

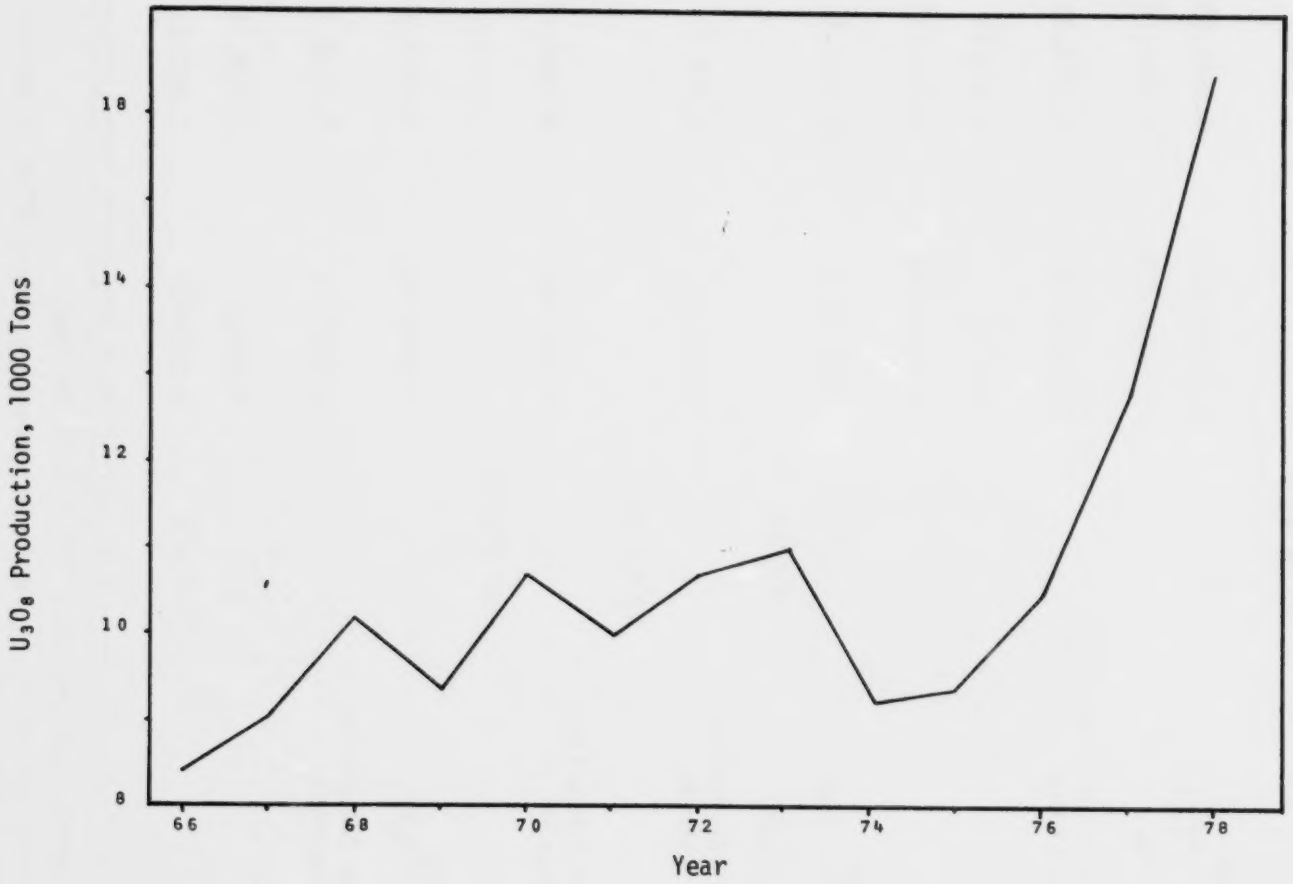

FIGURE 1. ANNUAL URANIUM CONCENTRATE PRODUCTION [12,29]

FIGURE 2. URANIUM MILLS IN THE UNITED STATES OPERATING AS OF JANUARY 1, 1979[29]

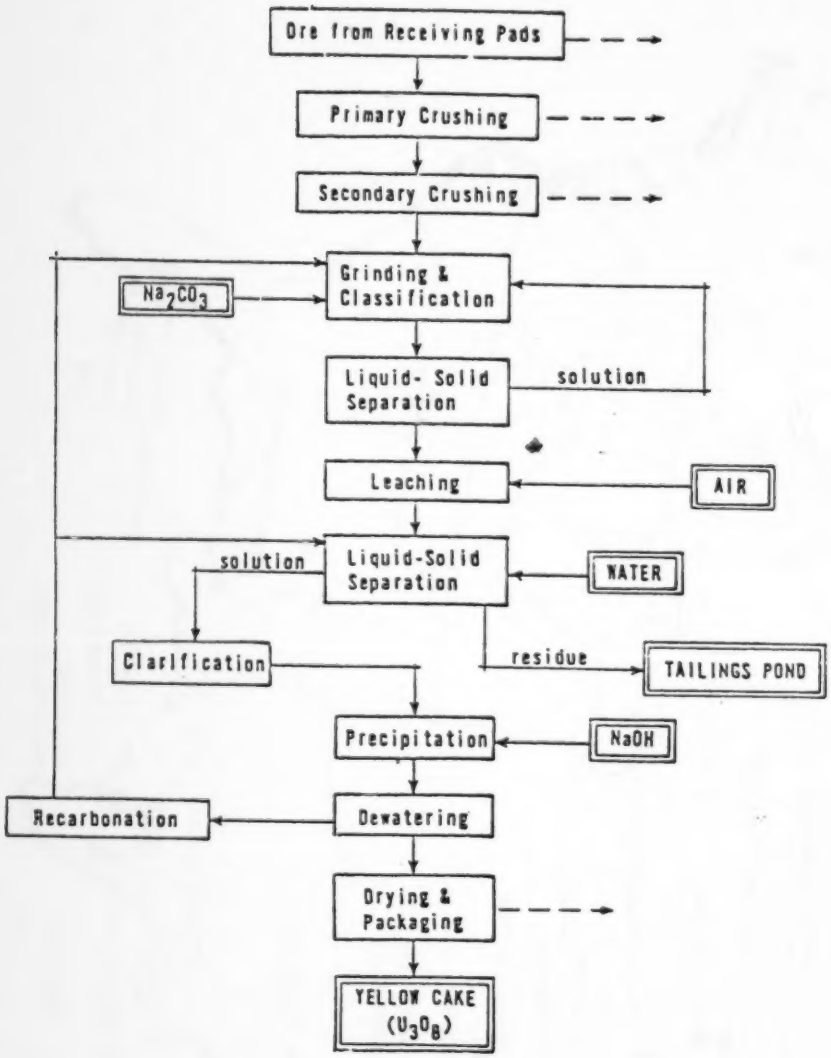

FIGURE 3. GENERALIZED FLOW CHART OF ALKALINE LEACH MILLING PROCESS[27]

LEGEND

--- Particulate Emissions
——— Process Flow

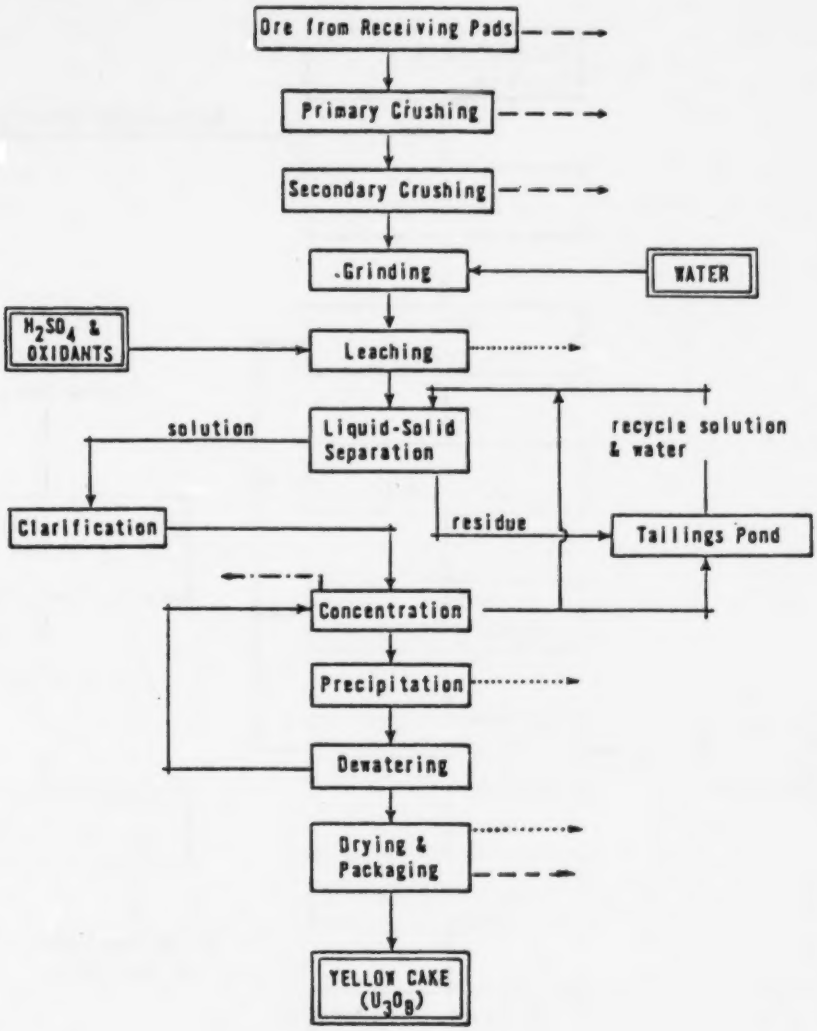

FIGURE 4. GENERALIZED FLOW CHART OF ACID LEACH MILLING PROCESS [27]

LEGEND

- – – Particulate Emissions
- ..... Acid Vapors, Ammonia
- —·—·— Hydrocarbons (Kerosene)
- ——— Process Flow

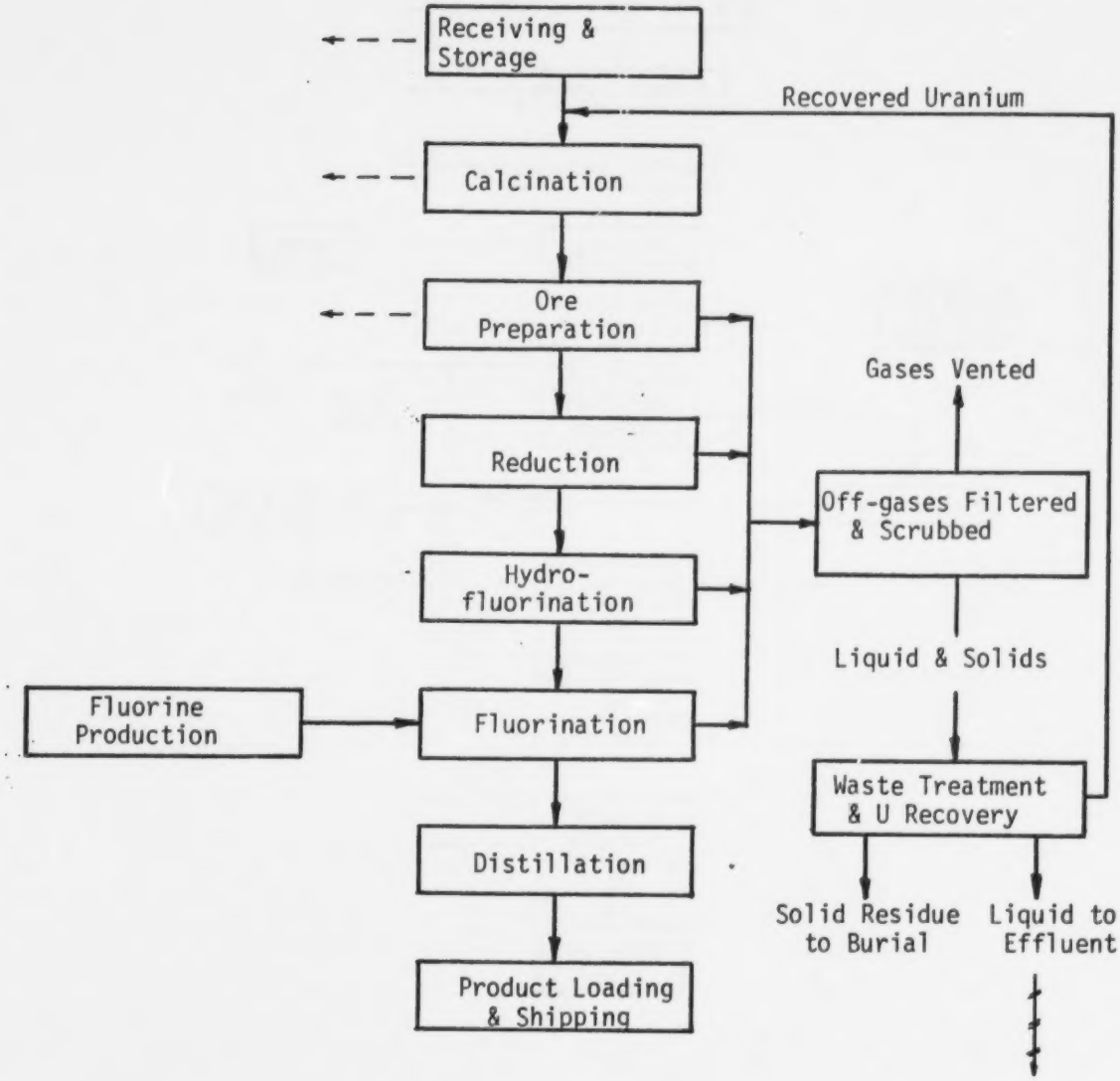

FIGURE 5. METROPOLIS (UF_6) FACILITY FLOW CHART[1]

LEGEND

- – – Particulate Emissions
- ~~~ Fluorides
- ——— Process Flow

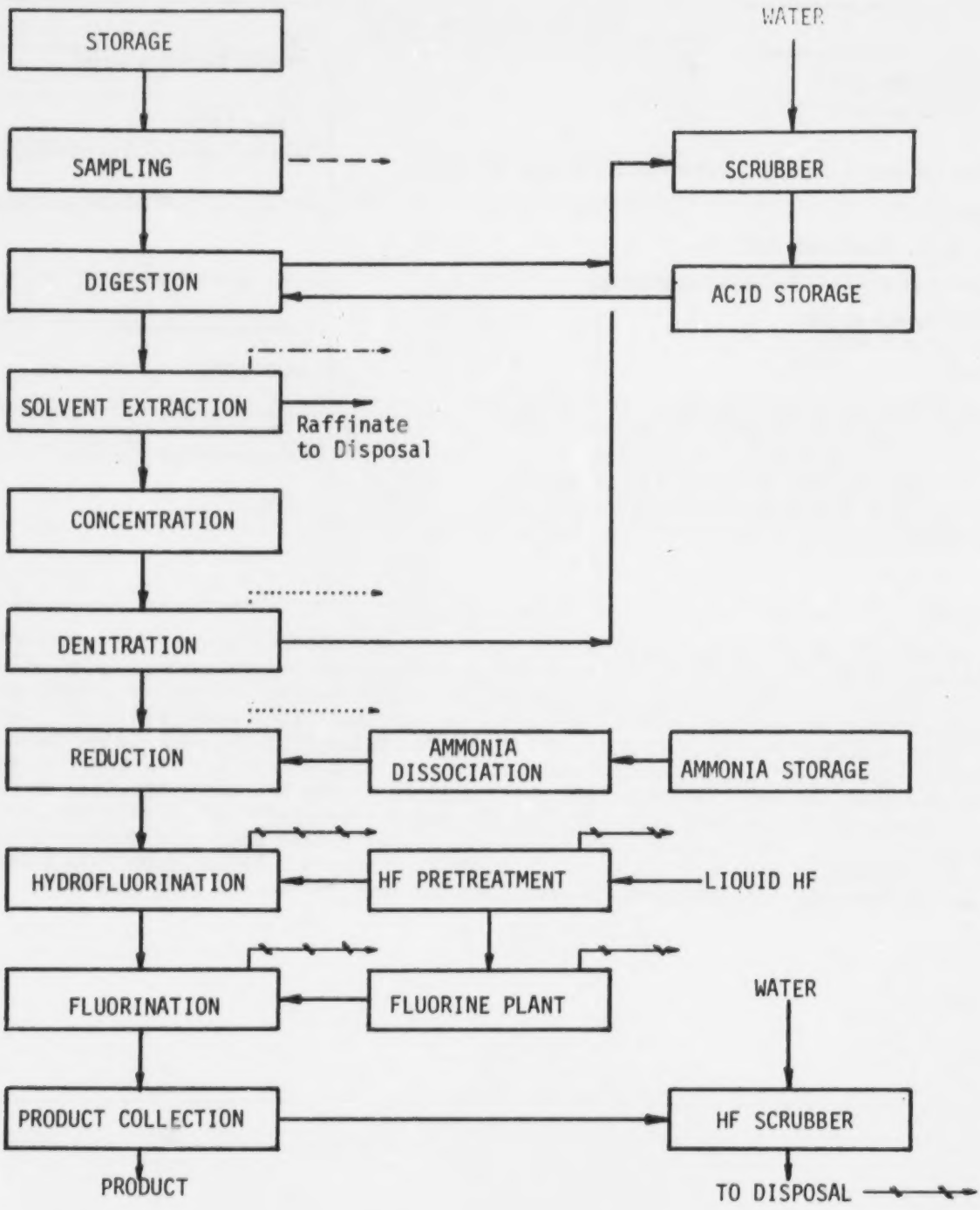

FIGURE 6. SEQUOYAH PLANT (UF_6) FLOW CHART [17]

LEGEND

- – – – Particulate Emissions
- ..... NO_x, Ammonia
- ~~~~ Fluorides
- –·–·– Hydrocarbon (Hexane)
- ——— Process Flow